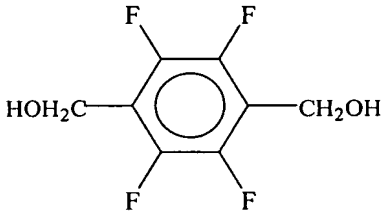
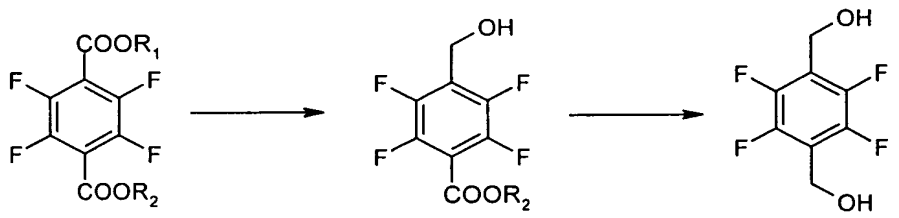
Claims

We claim:

1. A process for preparing 2,3,5,6-tetrafluorodimethylolbenzene, represented by the following structural formula, as one intermediate for pyrethroids,



which comprising a step of reducing dialkyl 2,3,5,6-tetrafluoro terephthalate in the presence of a reductant and a solvent according to the following scheme:



(I) (II) (III)

in the above formulae, R_1 and R_2 each independently represents a straight or branched alkyl chain having 1 to 6 carbon atoms, preferably, methyl, ethyl, n-propyl, n-butyl, n-pentyl, n-hexyl, iso-propyl, iso-butyl, tert-butyl or neopentyl.

2. The process according to claim 1, wherein both R_1 and R_2 represent methyl.

3. The process according to claim 1, wherein the solvent is selected from a group consisting of: alcohols; glycols; ethers; glycol ethers; glymes; polyglymes; polyethers; lower alcohols; two-phase solvent mixtures; polar inert solvents; organic acids; esters; water; ethers; lower anion surfactants or mixture thereof; or mixtures of two or more among them.

4. The process according to claim 3, wherein the solvent is methanol, ethanol, isopropanol, ethylene glycol, polyethylene glycols, diethyl ether, 1,2-dimethoxyethane, dioxane, tetrahydrofuran, diglyme or polyglyme, toluene, xylene, anisole, acetic acid, ethyl acetate, ethyl formate, water, tetrahydrofuran, or a mixture of two or more among them.

5. The process according to claim 3, wherein the solvent is methanol, ethanol, or a mixture thereof.

6. The process according to claim 1, wherein the reductant is a metal hydride, a borohydride, a metal aluminium hydride, aluminium borohydride, hydrogen or a hydrogen donor.

7. The process according to claim 6, wherein the borohydride is at least one selected from a group consisting of potassium borohydride, sodium borohydride, and lithium borohydride; and the metal aluminium hydride is lithium aluminium hydride.

8. The process according to claim 1, wherein the reductant is potassium borohydride, sodium borohydride or lithium borohydride and the process is carried out in the presence of a catalyst or an accelerant.

9. The process according to claim 8, wherein the accelerant is a denaturated metal salt or a boride.

10. The process according to claim 9, wherein the denaturated metal salt is one or more selected from a group consisting of aluminium, zinc, or titanium salts.

11. The process according to claim 10, wherein the denaturated metal salt is one or more selected from a group consisting of aluminium chloride, zinc chloride, and titanium tetrachloride.

12. The process according to claim 9, wherein the boride is boron trifluoride or alkyl boride.

13. The process according to claim 8, wherein the accelerant is lithium compound, preferably lithium chloride or lithium bromide, when using potassium borohydride or sodium borohydride as a reductant.

14. The process according to claim 8, wherein the molar ratio of the accelerant to the reductant is 0.05-1 : 1, preferably, 0.1-0.5 : 1.

15. The process according to claim 8, wherein the catalyst is onium salt, preferably one or more selected from a group consisting of tetra-alkyl ammonium salt, phosphonium salt, acyclic or cyclic-polyether.

16. The process according to claim 8, wherein the molar ratio of the catalyst to the reductant is 0.01-0.1 : 1.

17. The process according to claim 4, wherein the solvent is methanol, ethanol, isopropanol, ethylene glycol, polyethylene glycol, diethyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, diglyme or polyglyme.

18. The process according to claim 1, wherein the reduction is carried out using hydrogen as

the reductant in the presence of at least one catalyst selected from a group consisting of metals, metal oxides, mixed metal oxides, metal salts or metal complex catalysts.

19. The process according to claim 18, wherein the solvent is an alcohol, an aromatic hydrocarbon, an ether, an organic acid or ester thereof.

20. The process according to any one of preceding claims, wherein the process is carried out at from –20°C up to the boiling point of the solvent and is preferably carried out in the range of 30-120°C, more preferably in the range of 40-80°C.

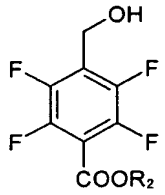
21. The process according to claim 1, wherein tefluthrin may readily be prepared by converting 2,3,5,6-tetrafluorodimethylolbenzene with the steps of halogenation, hydrogenation and esterification as follows:

i) halogenation of 2,3,5,6-tetrafluorodimethylolbenzene to give a 2,3,5,6-tetrafluoro-4-(halomethyl)benzyl alcohol;

ii) hydrogenation of the 2,3,5,6-tetrafluoro-4-(halomethyl)benzyl alcohol to give 4-methyl-2,3,5,6-tetrafluorobenzyl alcohol;

iii) esterification of 4-methyl-2,3,5,6-tetrafluorobenzyl alcohol with *cis*-((Z)-2-chloro-3,3,3-trifluoro-prop-1-enyl)-2,2-dimethylcyclopropane acyl chloride or *cis*-((Z)-2-chloro-3,3,3-trifluoro-prop-1-enyl)-2,2-dimethylcyclopropanecarboxylic acid to give tefluthrin.

22. An intermediate compound of formula (II),



(II)

wherein,

R_2 is a straight or branched alkyl chain having 1 to 6 carbon atoms and is preferably, methyl, ethyl, n-propyl, n-butyl, n-pentyl, n-hexyl, iso-propyl, iso-butyl, tert-butyl or neopentyl.

23. The compound according to claim 22, wherein R_2 is methyl.